Supporting Information

Formation of Hypoxanthine Tetrad by Reaction with Sodium Chloride: From Planar to Stereo

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1. Experimental details

All experiments were conducted in a ultrahigh vacuum chamber equipped with a variable-temperature Aarhus scanning tunneling microscopy (STM).[1] The typical base pressure was 1×10^{-10} Torr. The Au(111) single-crystal sample was cleaned by repeated cycles of 0.8 KeV Argon ion bombardment and annealed at 800 K for 15 min. Hypoxanthine (HX, Shanghai Aladdin Biochemical Technology Co., Ltd, 99%) was sublimated from a thermal evaporator heated to ~ 410 K and deposited onto clean Au(111) surface kept at room temperature (RT). All STM results were collected in the constant current mode at RT, with a bias voltage of 1.0-1.3 V and a tunneling current of 0.2-0.5 nA.

2. Calculation methods

Density functional theory (DFT) calculations were carried out using the Vienna *ab initio* simulation package (VASP).[2,3] The projector augmented wave (PAW) potentials were used to describe the interaction ionic cores and electrons and the Perdew-Burke-Ernzerhof (PBE) exchange-correlation density functional was used.[4] The dispersion interactions were considered within the dispersion-corrected DFT-D3 method of Grimme for the calculations when including the gold surface.[5] The force convergence criterion used for geometry relaxations was 0.02 eV/Å. The simulated STM images were based on the Tersoff-Hamann method. [6,7] The molecular clusters were optimized with the Au (111) substrate included. Partial charges were evaluated using Bader charge analysis based on DFT calculated electronic density.

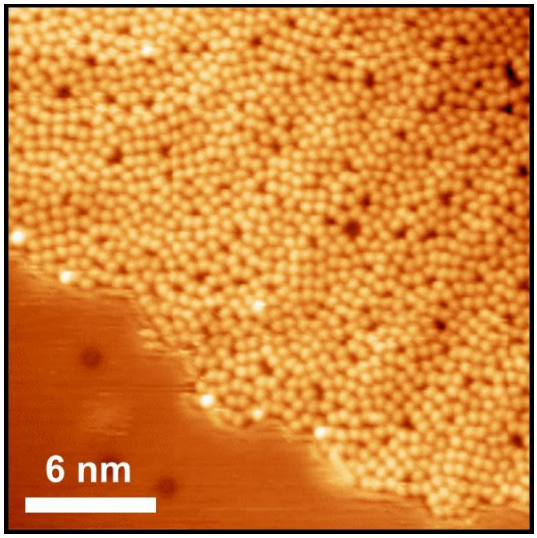

Figure S1. Disordered structure of HX molecules when depositing alone on Au(111) without post-annealing.

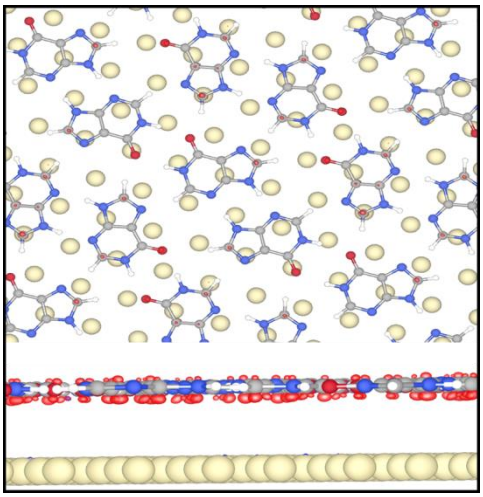

Figure S2. Top and side views of the electronic density of the close-packed structure of HX on Au(111) at the isosurface value of 0.006 eÅ^{-3}, showing no charge transfer with the substrate.

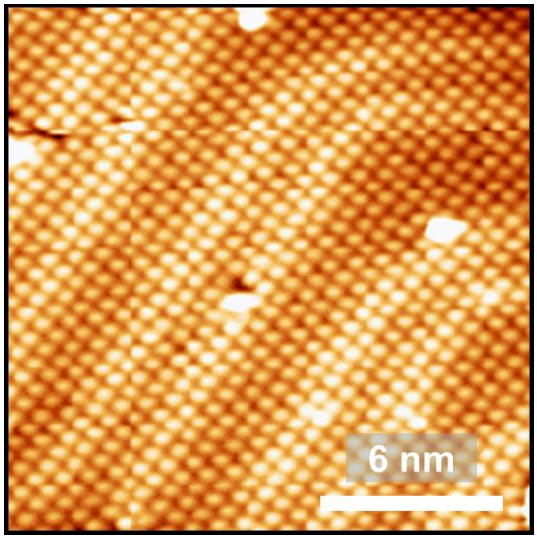

Figure S3. The close-packed structure of HX on Au(111), obtained after annealing, clearly showing a corrugation corresponding to the herringbone reconstruction of pristine clean Au(111) and hence indicating a weak HX-Au interaction in this case.

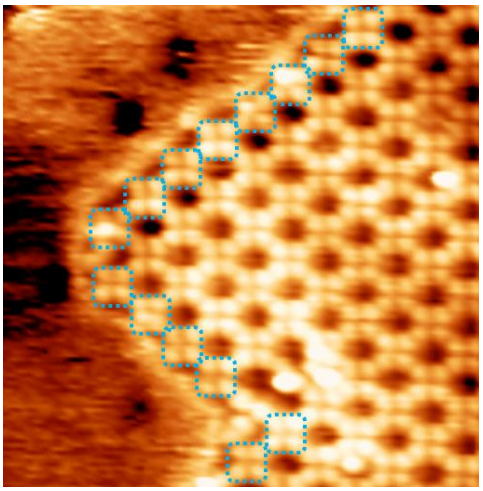

Figure S4. A 2-D domain of the planar HX-tetrads, where the edges of the networks are terminated by the tetrads (highlighted by dotted boxes).

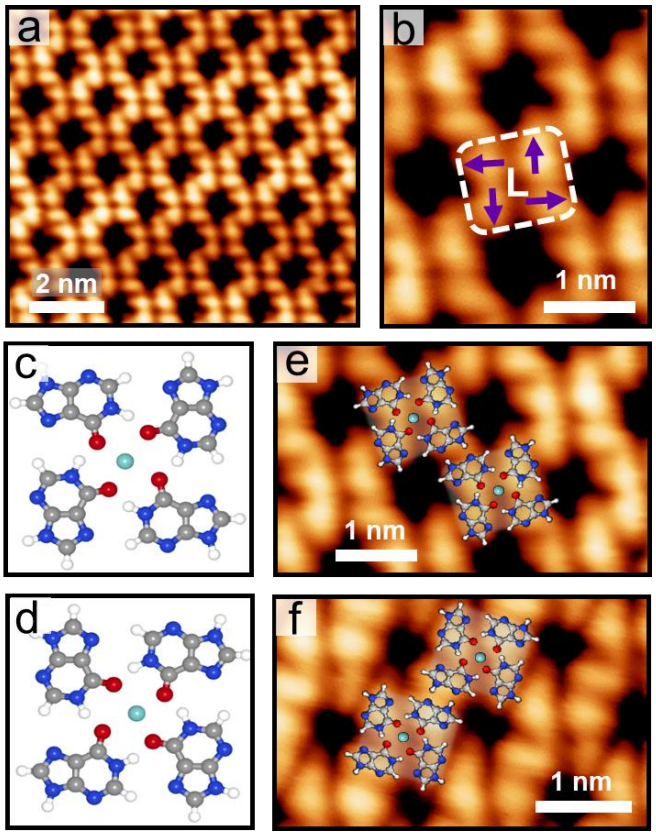

Figure S5. (a-b) Large-scale and zoom-in images of an anticlockwise arranged windmill-like tetramer network, with each tetramer comprised of four L-chiral HX. (c-d) Model of the two homochiral tetrads composed by L- and D-chiral HXs and one Na in the tetrad center. (e-f) Close view of the networks of L- and D-tetrads superimposed with the calculated model.

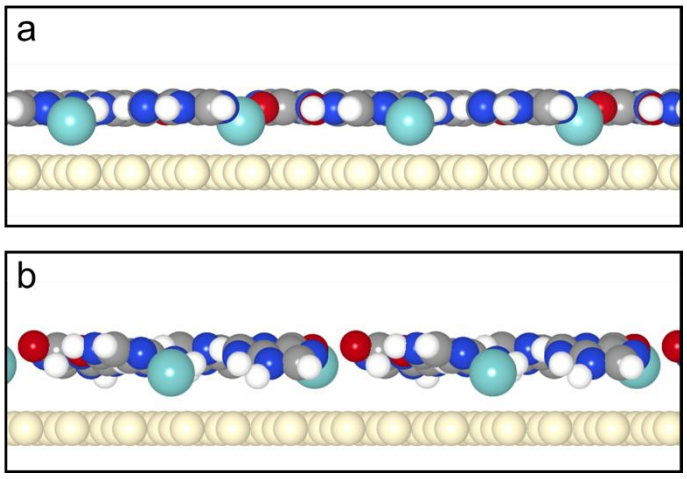

Figure S6. (a) Starting planar configuration and (b) relaxed structure of the calculated non-planar HX-tetrads with Na on the Au(111) surface.

References

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